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THE GROWTH OF LARGE DIAMETER SINGLE CRYSTALS BY
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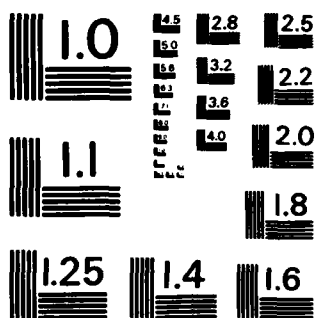
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THE GROWTH OF LARGE DIAMETER SINGLE CRYSTALS BY VERTICAL SOLIDIFICATION OF THE MELT

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CERAMICS RESEARCH DIVISION

August 1984

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ABSTRACT

↓ Growth of large diameter single crystals, especially of phases which melt at high temperatures and have a high specific gravity, is governed by conditions that quite differ from those which occur during the growth of crystals whose diameter does not exceed 50 mm. This paper defines and analyzes these special conditions and delineates their effect on crystal growth. It is also shown that in spite of the presence of negative factors which are inherent to the growth process of large diameter single crystals, these adverse factors, if recognized properly, can be brought under control to such a degree that the growth of large diameter single crystals can be achieved. The vertical solidification of melt method (VSOM) is described. This method produces large single crystals, even for binary mixtures with a steep liquidus. Neodymium-doped yttrium aluminum garnet (Nd:YAG) crystals have been grown 75 mm in diameter and 100 mm high. These crystals are also water clear. Both size and perfect crystallinity are important for application to lasers. ↗

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INTRODUCTION

In 1918, Czochralski¹ developed a method for measuring speed of crystallization of metals. This technique soon found its general use in crystal growth, at first in the formation of wire-like single crystals and later in the production of a great majority of single crystals grown from the melt. However, there are some limiting factors in Czochralski's technique, which prevent its application to the growth of large diameter single crystals, particularly from materials which simultaneously have a high melting point and high specific gravity, and are also multi-component systems.

In 1924, Ströber² introduced a technique which was based on cooling a melt confined in a hemispherical dish by resting it on a water-cooled heat sink. This process is most suitable for the growth of single crystals which possess a unique crystallographic axis along which much greater heat conductivity is associated than in other directions. The disadvantage of this technique is that the loss of a thermal gradient towards the end of solidification causes an uncontrollable growth, resulting in low quality crystals.

In 1974, Viechnicki and Schmid³ overcame the disadvantages of the latter method by controlling both the nucleation and loss of the thermal gradient, thus enabling growth of large single crystals. However, this Heat Exchanger Method (HEM) proves to be useful for growing large diameter single crystals mainly from single component systems.

Recently, in our laboratory a method was developed for growing laser quality single crystals from materials belonging to binary or even to more complicated systems. This technique is called the Vertical Solidification of the Melt (VSOM), because the solidification commences at the bottom of the crucible containing the melt. Advantages as well as problems associated with the VSOM technique will be analyzed in this paper.

STATEMENT OF THE PROBLEM

Production of single crystals from the melt involves an arrangement of conditions in which atoms of the melt "freeze out" on a nucleus or seed crystal and perpetuate the lattice array of the nucleus.

1. CZOCHRALSKI, J. Zeits. Physics. & Chem., v. 8, 1918, p. 219.

2. STRÖBER, F. Zeits. f. Kryst., v. 61, 1925, p. 299.

3. STRONG, J. Phys. Rev., v. 36, 1930, p. 1663.

If the crystal is to be grown at the expense of the liquid, whose temperature is at all points above the melting point, then the latent heat can only be removed by conduction down through the already formed crystal. Thus, a temperature gradient must be maintained along the length of the crystallized solid.

In the majority of the crystal growth techniques, the propagation of the solid-liquid interface is achieved by drawing the crystal through a thermal gradient or by moving the furnace itself. This motion causes vibrations, and thermal fluctuations which may deleteriously affect the growth of crystals, especially large diameter crystals destined for laser purposes.

As previously mentioned, the Czochralski method is not well suited for the growth of large diameter, laser quality single crystals from materials of high specific gravity and high melting temperature. These two factors combined, especially in the presence of vibrations caused by pullers, increase the probability of stress occurrence in the neck of the crystal. Generally such stress is a leading source of crack nucleation. Furthermore, thermal fluctuations during the growth can cause defects to form in large diameter single crystals.

The Ströber technique eliminates the problem of vibrations by simply moving the thermal field through the system instead by moving the crucible or the furnace. Its mode of cooling is designed in such a way that the solidification commences at the bottom of the container and proceeds upwards. The upward thermal gradient has a stabilizing effect on the growth, but in order to eliminate the polycrystalline growth, the nucleation must start from a single nucleus, i.e., from almost a point. In 1930, using the Ströber technique, Strong⁴ grew 100 X 100 mm alkali halides crystals; however, at the end of the process, he lost growth control due to an insufficient thermal gradient which caused the upper portion of the crystal to be cloudy.

Viechnicki and Schmid³ overcame both the nucleation problem and loss of the thermal gradient by placing the seed crystal above a helium-cooled heat exchanger. Cylindrical sapphire single crystals were grown, 130 mm high, 250 mm in diameter, with a low density of dislocations and almost

4. VIECHNICKI, D., and SCHMID, F. *Crystal Growth Using the Heat Exchanger Method (HEM)*. J. Crystal Growth, v. 26, 1974, p. 162.

free of grain boundaries.⁵ The Heat Exchanger Method (HEM), however, did not prove to be suitable for the growth of large diameter laser quality single crystals of compositions which belong to binary systems, with a steep liquidus. The reason for this unsuitability seems to be associated with principles which, unfortunately, are inherent in the technique. In the HEM, the melt is kept at constant temperature while growth of the crystal is controlled by the thermal gradient within the solid, and directed by the helium flow in the heat exchanger. Since the temperature of the solid-liquid interface is constant at any given point, the melt temperature is not influenced for all practical purposes either; therefore, it remains uniform in almost the entire volume of the melt. In order to sustain the growth, the thermal gradient in the solid has to be continuously increased. At the beginning of the process, the melt has a comparatively large volume while the crystal growth has to originate from a relatively small seed crystal. In a homogeneous medium, heat propagates in a straight path. Therefore, the solid-liquid interface at the beginning of the solidification takes a hemispherical shape, and consequently the growth becomes three dimensional. The difference in shapes of the hemispherical interface and of the melt, which is contained in a cylindrical crucible, causes the growth rate to vary in the melt due to differences ($\Delta T/l$) in all directions perpendicular to the tangents drawn at any given point of the interface. (For the definition of $\Delta T/l$, see Figure 1.) Hence, as the growth progresses, curvature of the solid-liquid interface becomes more and more convex, i.e., takes on a paraboloid-like shape which leads to a formation of bizarre pear-shaped interfaces in the last stages of the solidification process (Figure 2).

NOTE: The liquidus of two different congruently melting compounds, in the close proximity of the compounds' melting point, may have an entirely different character. Figures 3a and 3b depict two rather extreme types of liquidus curves. Compounds with a melting behavior described by the type of liquidus shown in Figure 3a, and naturally those of single component systems, are easily grown as single crystals, since only minor temperature adjustments are required during the growth, even if significant composition changes occur in the melt. On the other hand, growth of single crystals

from a melt with compositions characterized by a liquidus shown in Figure 3b, is strongly dependent on the equilibrium temperature. Hence, even small deviations of the melt composition would require major readjustments of the growth temperature.

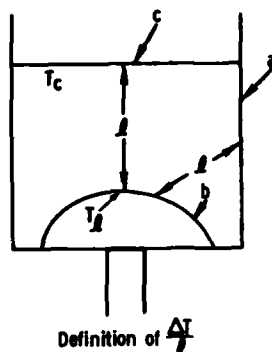


Figure 1. Shows melt confined in cylindrical crucible a; convex solid-liquid interface b; and melt surface c. T_i is a temperature of solid-liquid interface T_c is the temperature at the surface of the melt and l is a perpendicular drawn to tangent at any point of solid liquid interface, hence $(T_c - T_i)/l = (\Delta T/l)$.

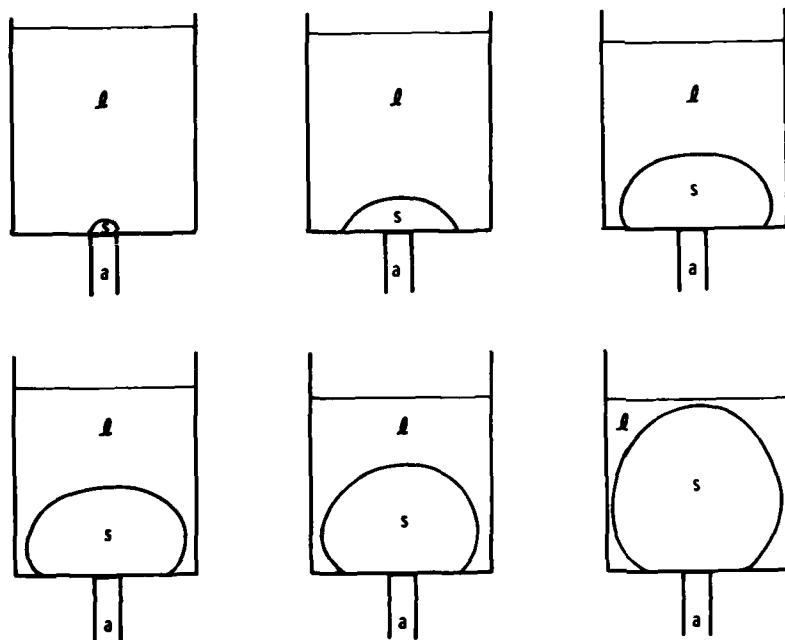


Figure 2. Change of the shape of the solid-liquid interface with progressing growth in HEM technique; l = melt, s = solid, and a = helium heat exchanger.

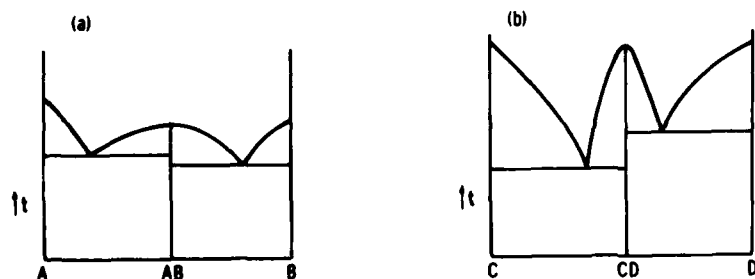


Figure 3. Liquidus curves depicting extremes in melting differences of two hypothetical binary compounds.

Crack Formation in Large Diameter Single Crystals

Until now the discussion of problems associated with the growth of large diameter single crystals was focused on employing the well-established growth techniques and adapting them for a scale-up of crystal diameter. Yet, the problems may also have other origins which may or may not be solely inherent in the techniques of growth. They may be traced, rather, to the specific conditions and mechanisms of the growth of large diameter crystals, or to origins which even have not been sufficiently identified. In spite of that, if the problem is correctly recognized and conditions of the growth amended, large diameter crystals may be grown successfully, as is documented in the case of growth of large sapphire single crystals.

Thermal oscillations generally affect the quality of crystals of any size. If we take a system in which the solid-liquid interface has a convex shape to the melt, then, due to the anisotropy of the growth directions, the interface will take on a faceted shape, as seen in Figure 4. These facets form pockets capable of trapping inclusions which can be permanently imbedded in the crystal. An interface of an incongruent particle in the crystal matrix can accept or emit point defects. At a given temperature, the crystal matrix contains an equilibrium concentration of point defects.



Figure 4. Optical photograph of decanted interface of 200 μm diameter sapphire crystal showing pockets formed by facets. The $(10\bar{1}2)$ plane is in the plane of the photograph.

On changing the temperature, this concentration will arrive at a new equilibrium value by drawing point defects from or depositing them into the interface between the matrix and the particle. This results in a change in volume of the hole containing the inclusion, leaving the size of the particle itself unchanged. The misfit thereby created, can develop stresses large enough to nucleate prismatic loops around the particle. If during the crystal growth temperature cycles, a procession or processions of prismatic loops develops in the crystal matrix (see Figure 5a), the accompanying stress to each of the individual loops of the procession is added consonantly. If the resolved value of the emanating stress from the procession of the loops exceeds binding forces of a weak crystallographic plane, then a crack may nucleate in the growing crystal (Figure 5b). This type of crack formation is of a general nature and can be found regardless of the size of the crystal diameter.

Another factor which may lead to crack formation is concentration of stresses in the area of the crystal's neck by the crystal's weight. This stress may occur under conditions which are typical for the Czochralski method, namely, when large diameter single crystals of a high specific gravity are grown. The critical period occurs after the seed is dipped into the melt and the neck of the crystal is formed. To assure the single crystallinity of the growth, the neck diameter has to be increased very rapidly to the final diameter of the crystal. With increasing diameter, the weight of the crystal increases with r^2 , so that at some point the crystal's weight will set stress in the neck of the crystal. This stress activates the dislocation sources in the growing crystal. In the case of a large diameter single crystal, numerous dislocation sources are activated simultaneously, causing formation of dislocation tangles⁵ (Figure 6). Under stress, the edge dislocation will glide (see Figure 7), but in specific directions only. Due to this glide, edge dislocations form tilt grain boundaries in the crystal,⁶ where the magnitude of the tilt of the grain boundary is indirectly proportional to the spacings between the dislocations and, therefore, also to the stress. Hence, magnitude of the tilt of a grain boundary formed by this mechanism is a function of the

5. CASLAVSKY, J. L., GAZZARA, C. P., and MIDDLETON, R. M. *The Study of Basal Dislocations in Sapphire*. *Phil. Mag.*, v. 25, 1972, p. 35.

6. CASLAVSKY, J. L., and GAZZARA, C. P. *Dislocations Behaviour in Sapphire Single Crystals*. *Phil. Mag.*, v. 26, 1972, p. 961.

stress in the neck of the crystal, and thus it depends on the crystal's weight, which is proportional to the specific gravity of the material. In other words, the frequency of the grain boundary formation, as well as



Figure 5. X-ray topographs of two incoherent inclusions [indicated by arrows in plate (b)] in sapphire single crystal. Plate (a) shows two processions of prismatic loops. In plate (b) loops are in extinction; they nucleated crack indicated by arrow in plate (a). $\text{Ag } K\alpha_1$ radiation.



Figure 6. X-ray topograph of dislocation tangles in sapphire single crystal. Note a formation of low-angle grain boundaries. Ag $K\alpha_1$ radiation.

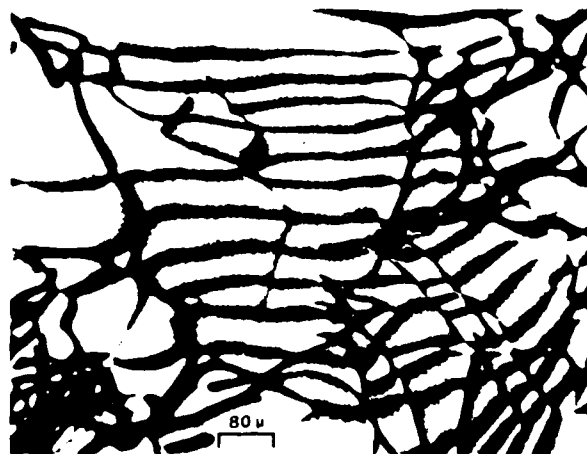


Figure 7. X-ray topograph of edge dislocations gliding under the stress in a sapphire single crystal. Ag $K\alpha_1$ radiation.

the magnitude of the grain boundary angle, is directly proportional to the rate with which the crystal diameter increases from the neck region. Based on experimental data obtained during the growth of large diameter single crystals of refractory oxides, it was found that if the grain boundary is perpendicular to the stress vector, and the tilt angle of the boundary exceeds 7° , then there is a 50% probability that the grain boundary will nucleate a crack during the plastic brittle transformation. The higher the tilt angle, the higher is the probability of crack nucleation. Since the probability of formation of a high angle boundary increases with the increase in the diameter, this mechanism of crack nucleation is likely to occur under the conditions of the Czochralski method, or any other method where the crystal "hangs by its neck." In other words, since the crack nucleation is directly correlated with the crystal's weight, by the Czochralski method, there is a lesser probability of crack formation in growing large diameter silicon single crystals than in growing sapphire crystals of the same size, in spite of the fact that both materials belong to single component systems. Hence, if growth of large diameter single crystals is plagued by the formation of cracks, a detailed study of dislocation configurations is recommended in order to find out if the reason is in the choice of the method or in an inadequate temperature control or some other factor.

Second Phase Segregation

Segregation of a second phase generally occurs during the crystal growth due to constitutional supercooling. (In laser crystals, second phase particles are called scattering centers.) This phenomenon can be caused either by impurities or by any material whose composition belongs to a binary system with a steep liquidus. In the former case, the remedy is easily at hand; in the latter case, nucleation of second phase particles is frequently traced to the curvature of the solid-liquid interface. Figures 8 and 9 are photographs of cross sections of sapphire and 1.0 At% Nd:YAG single crystals respectively, both crystals grown by the HEM technique. A highly curved interface is seen in both crystals, but there is an absence of scattering centers in the sapphire crystal, while a high density of second phase particles is deposited in the Nd:YAG crystal. Since both materials have a high melting point, it was possible to grow both types of the crystals under almost identical conditions. The difference in the crystal quality is because sapphire is a single component refractory oxide, while YAG is a binary compound of refractory oxides, whose melting behavior is characterized by a steep liquidus. If such a compound is grown on a curved interface inherent in the HEM technique, variations of $\Delta T/l$ in the melt will cause constitutional supercooling and consequently growth instability. Constitutional supercooling is relieved by a compositional change in the liquid resulting in a deposition of second phase particles



Figure 8. Optical photograph of a cross section of sapphire single crystal. Before the completion of solidification the remaining melt was decanted to reveal the shape of solid-liquid interface. Crystal grown by HEM.



Figure 9. Optical photograph of the cross section of a 1 At% Nd:YAG single crystal grown by HEM technique.

along the solid-liquid interface. After the constitutional supercooling is eliminated, the controlled growth is restored. However, the process will repeat itself, leaving alternating clear and translucent bands in the crystal (Figure 9). The geometrical conditions in the HEM apparatus were rearranged to simulate VSOM to enable the Nd:YAG crystals to be grown on a planar interface. The optical photograph in Figure 10 shows a vertical cross section through such a crystal. This photograph shows that the crystal grows from an almost planar interface. There are no layers formed due to the constitutional supercooling. The presence of second phase particles observed in this case and the mechanism of their formation were described by Caslavsky and Viechnicki in 1980.⁷

Planar solid-liquid interface is essential for growing good quality single crystals. But as shown experimentally, it is difficult to maintain it during the entire period of the growth of a large diameter single crystal.

Hence, in order to grow the large diameter, laser quality single crystals from a material which simultaneously comprises all of the properties discussed above, one has to select a growth technique which would enable the following:

- a. support of the growing crystal preferably along its entire diameter (rather than having crystal hanging from a narrow seed);



Figure 10. Optical photograph of the cross section of a 1 At% Nd:YAG single crystal grown by VSOM technique.

7. CASLAVSKY, J. L., and VIECHNICKI, D. *Melting Behaviour and Metastability of Yttrium Aluminum Garnet (YAG) and $YAlO_3$ Determined by Optical Differential Thermal Analysis.* J. Mater. Sci., v. 15, no. 7, 1980, p. 1709.

- b. determination of the crystal diameter by the container shape rather than by balancing the heat flow through the system;
- c. elimination of mechanical vibrations by application of a mobile thermal field instead of moving the furnace or the crucible or the crystal itself.

Such a crystal growth technique, capable of growing large diameter single crystals from even complicated melts, was developed in our laboratory, and has been named the "Vertical Solidification of the Melt" (VSOM). The method is examined in detail in this paper.

CONTROL OF THE FACTORS AFFECTING THE GROWTH OF LARGE DIAMETER SINGLE CRYSTALS

Previous paragraphs have discussed parameters with a negative influence on the growth of large diameter single crystals. A thorough analysis of these factors revealed that they are intrinsic to the process; hence they can not be eliminated, but their negative effect can be minimized by a combination of compensating conditions. This leads to the creation of a new crystal growth technique which maximizes the potential for growing large diameter single crystals from melts of a wide variety of compositions. However, to ensure smooth operation of the technique, the following conditions must be concurrent and co-operative, and must be observed so that they are under control all the time:

1. The growing crystal must be supported on its entire cross section.
2. Mechanical vibrations must be eliminated.
3. The solid-liquid interface has to become planar on the macroscopic scale, commencing at a specific point, and must remain so until the end of solidification. Accordingly, the Vertical Solidification of the Melt method was developed in our laboratory, VSOM implies the method's main feature, i.e., to respect the nature of the direction of solidification. An apparatus for the crystal growth by vertical solidification was designed and built at the Army Materials and Mechanics Research Center.

At this point, it is proper to discuss separately all the crucial factors which contributed to the design of this apparatus then the analysis and resolution of these factors into the new technique.

The easiest requirement to fulfill was that of supporting the entire cross section of the growing crystal. The material to be solidified was simply placed in a cylindrical container with a diameter of the required crystal size. To commence solidification at a single point and in a

single crystalline mode, a seed crystal was placed in the center of the bottom of the container. The entire charge of the material had to be melted first; hence, in order to protect the seed crystal from melting, the crucible was rested on a cold finger which kept the seed crystal below melting temperature, hence, preventing melting. Then during the period of shaping the solid-liquid interface, and especially during the entire period of the crystal growth, this cold finger takes on the function of a heat sink, thus having a crucial importance for controlling the crystal growth.

Mechanical vibrations were eliminated by the formation of a suitable heat zone which imposes on the solid as well as on the liquid, a thermal field whose isotherms must be parallel with the bottom of the crucible and have an upward gradient. In order to obtain the driving force for the crystal growth, the thermal field is propagated through the system in the desired fashion and rate. This required function is derived from two optical automatic pyrometers (#1 and #2 as illustrated on Figure 11). The basic idea of the design is as follows: pyrometer #1 senses the temperature of the black body and thus controls the desired temperature of the whole system. Pyrometer #2 observes the temperature of the content of the crucible, i.e., it controls the temperature of the melt. In order to start the growth, the temperature of the black body is slightly lowered. Since the heat capacity of the content of the crucible is significantly higher than the black body heat capacity, a temperature difference occurs between the black body and the melt temperature. This difference is seen by optical pyrometers, translated to an appropriate electrical signal and fed into a microprocessor whose program is designed to control the temperature decrease of the black body as a function of ΔT . In this case ΔT is a difference between the temperature of the black body and the surface of the melt. In other words, the rate of temperature decrease of the black body is controlled by the program as a function of the temperature difference between the crucible content and black body. This has a crucial importance. During solidification of crystals with large cross section, a significant amount of a latent heat is developed on the interface; hence, this heat influences the melt temperature. This change is sensed by pyrometer #2, whose signal in turn corrects the signal of pyrometer #1. This type of control enables programming the propagation of the thermal field with

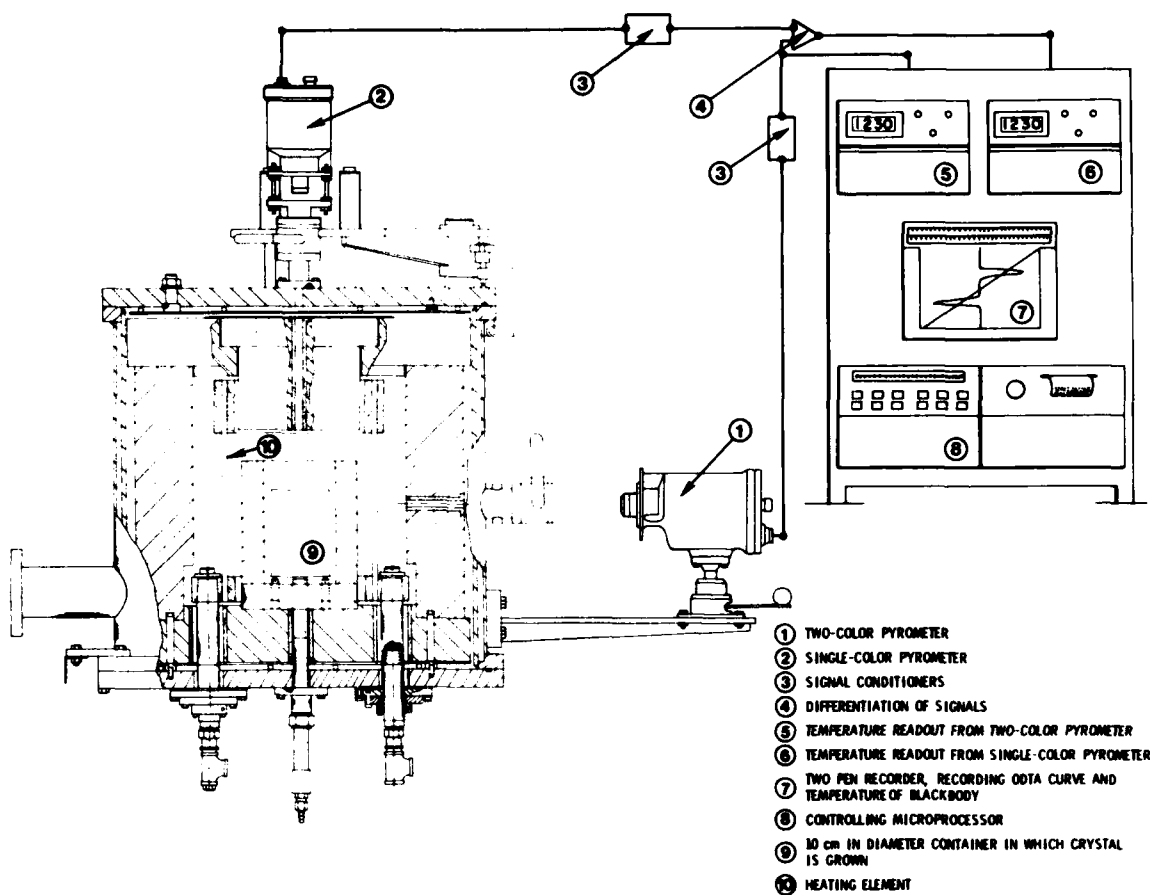


Figure 11. Schematic of crystal growth furnace for vertical solidification of the melt (VSOM).

an accuracy of $\pm 1^\circ\text{C}$ in the temperature range between 1600°C to 2100°C , and the rates of temperature decrease between 2.0 and 0.1°C/h ; i.e., the temperature of the thermal field, measured at any point along the vertical axis of the field, can be controlled with an accuracy $\pm 1^\circ\text{C}$ in the given range, and the temperature at such a point of the thermal field can be either decreased or increased as slowly as 0.1°C/h linearly in the given range. In principle, this is the driving force for the crystal growth--a very accurate control. However, to keep isotherms of the upward thermal field mutually parallel, the latent heat of solidification has to be removed from the system down through the already crystallized solid and down into the heat sink. Therefore, in order to grow a single crystal by vertical solidification and in a controlled manner on a flat interface, the heat flow through the heat sink has to be equal to the net heat flow through the interface conducted down through the already crystallized charge. The heat transfer equation describing the heat flow through the interface and the solid may be written as follows:

$$K_s A_s \frac{dT}{dx_s} - K_l A_l \frac{dT}{dx_l} = AL\rho \frac{dx}{dt} \quad (1)$$

where: K = thermal conductivity

A = cross section area of the charge

T = temperature

x = a coordinate measured along the vertical axis of the system

ρ = density of the material

L = latent heat of the crystallizing material (suffixes s , l and h relate to the solid and liquid phase and to the heat sink respectively see graphical illustration in Figure 12).

Since the cross section A_s and A_l and the interface A are approximately equal, we may write that:

$$K_s G_s - K_l G_l = RL\rho \quad (2)$$

where: G = temperature gradient

R = growth rate.

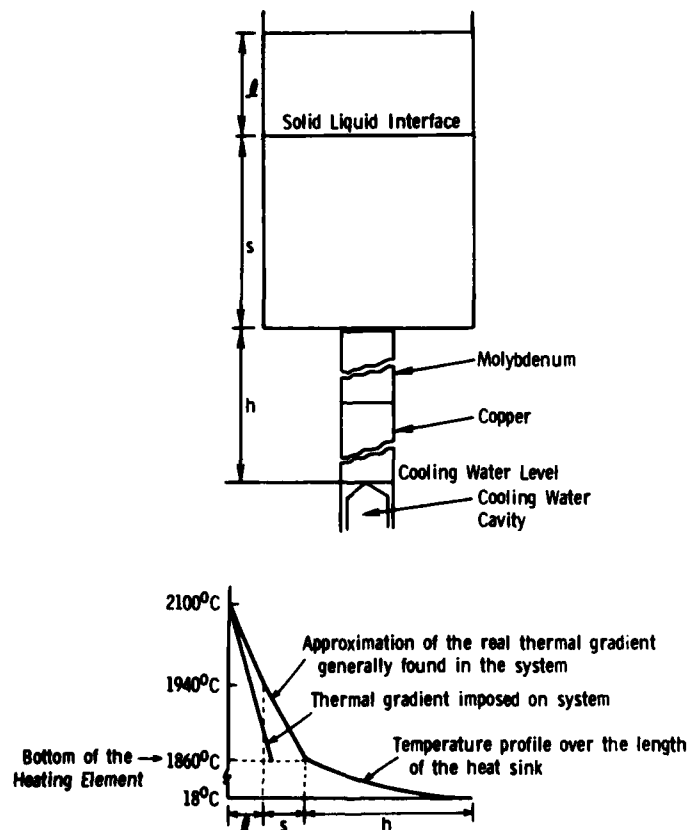


Figure 12. Graphic illustration of cross section of the crucible and the heat sink.

In this case, the growth rate is the movement of the interface and the rate of the growth of the crystal is governed by the rate at which the latent heat is extracted from the system by the heat sink. The heat flow ϕ through the heat sink can be expressed:

$$\phi = K_h A_h \frac{dT}{dx_h} \quad (3)$$

and in the first approximation:

$$RL\rho = K_h A_h \frac{dT}{dx_h} \quad (4)$$

Because of complicated boundary conditions and the practical impossibility of determining the exact position of the interface, Equation 4 cannot be solved numerically. Therefore, ϕ has to be determined experimentally, from the grown crystal. If the heat sink removes an insufficient amount of heat, i.e., if

$$RL\rho > K_h A_h \frac{dT}{dx_h}, \quad (5)$$

then a dendritic growth is observed (see Figure 13). Or if

$$RL\rho < K_h A_h \frac{dT}{dx_h}, \quad (6)$$

i.e., if the heat sink removes an excessive amount of heat, the interface will become convex (see Figures 8 and 9).



Figure 13. Optical micrograph of decanted interface of sapphire crystal grown by VSOM technique under $RL\rho > K_h A_h (dT/dx_h)$ condition. $[0001]$ directions is in the axis of the sapphire dendrite. The crystal was grown in $[5052]$ direction perpendicular to the plane of photograph.

Once the direction of an error is known, the heat flow capacity of the heat sink can be corrected in the following way. Designing of the heat sink for growing large diameter crystals with a melting point above 1600°C is influenced by two important factors. Total heat flow in the heat sink is considerable; hence, it has to be made from a material with high thermal conductivity. Copper is a material with high heat conductivity, but copper is not practical with high temperatures because its melting point is low (1083°C). Nevertheless, the main body of the heat sink (#2, Figure 14) is designed to be made of copper, but it is extended by tungsten to the region of the high temperature (#1, Figure 14). The dimensions of the surface are determined by diameter "f" (Figure 14) and its optimal value was found experimentally to be 1/15 of the interface surface area. Value of "a" has to be determined by trial and error. To approach closely Equation 4, the fine tuning of the ϕ value can be achieved by adjusting dimensions "c" and "e" (Figure 14), to a degree that the heat flux ϕ required for steady growth can be controlled only by moderating the flow of the cooling water through the heat sink. This is done at 50% of maximum cooling water flow at the water temperature 2°C higher than the water temperature required during the growth.

In our laboratory we have focused on growing large diameter crystals of refractory oxides with the melting point in the 1800° to 2100°C range. For this reason, resistively heated graphite furnaces are used. To protect the graphite and the heat sink including the molybdenum crucible, a low vacuum is used, approximately 5 times 10^{-2} . At first conventional "ribbon" type element was used, but electric current in that type of element (Figure 15 on the right hand side) is prone to develop alternating hot and cold spots which effect circumferential and axial temperature gradient. Such an element, therefore, proved useless for the growth of crystals by the vertical solidification of the melt. In 1976, a graphite element called a "bird cage" was designed and built (Figure 15, left). This element consisted of a complete bottom ring, a split upper ring and forty tapered graphite rods connecting the rings. These rods are tapered proportionally to the required upward thermal gradient. Originally, in the first design of the element, the power feeds were attached to the upper element ring. Because of the heat developed by the element, and also because of the need to operate the

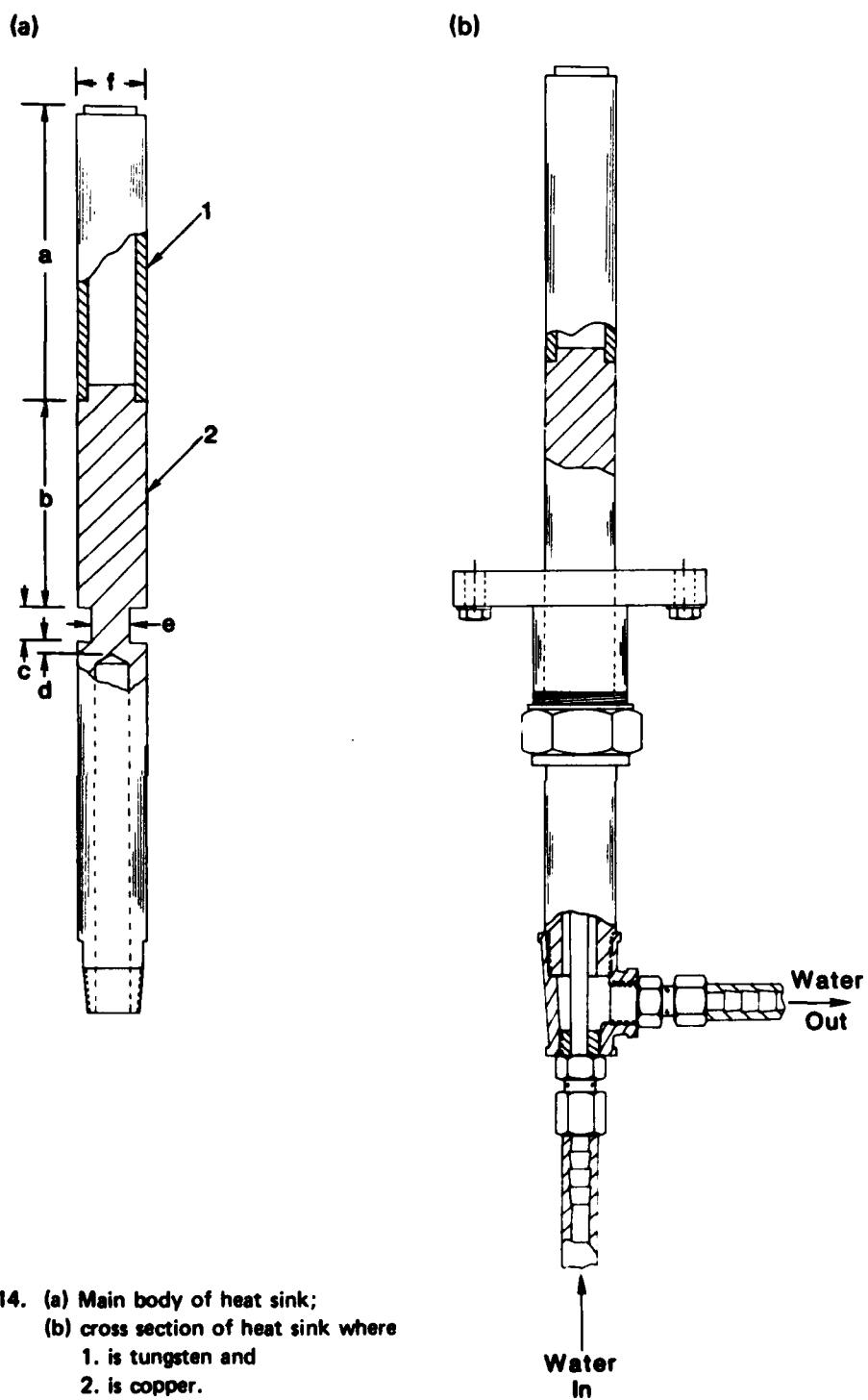


Figure 14. (a) Main body of heat sink;
(b) cross section of heat sink where
1. is tungsten and
2. is copper.



Figure 15. Photograph of graphite heating elements. Conventional ribbon element on the right side of the photograph. Note the thermal pitting and hot and cold spots alternation on the ribbon element. "Bird Cage" element on the left side of the photograph.

element at high temperatures requiring a high electric current, the power feeds have to be rigorously cooled by water. Consequently, they act as powerful heat sinks, thereby causing a decrease of the vertical thermal gradient. To remedy this problem, an element was inverted so that the power feeds were brought to the lower element ring. In this design, a sufficient upward gradient of $1.0^{\circ}/\text{mm}$ over the 120-mm element heights was achieved. This element was adequate to enable growth of Nd:YAG single crystals 75 mm in diameter and 100 mm high. Since the gradient thus achieved proved to be still insufficient for growth of YAG crystals with a 100-mm diameter, a new type of "bird cage" element was designed and built. Because this element design has been submitted for patent proceedings and the patent is pending, no details can be given at the present time. This is the reason why the term "an upward thermal gradient" describing the thermal field imposed on the crucible containing both the growing crystal and the unused melt was previously introduced without any further comments.

CONCLUSIONS

Factors having a negative influence on the growth of large diameter single crystals were discussed in detail. Analysis of these factors revealed that the degree of their adverse action on crystal growth can be evaluated, and therefore their influence minimized, by imposing upon them a set of suitable ballancing conditions. Recognition of this principle led to a design of a new crystal growth technique (VSCOM) which allows growth of single crystals of certain compositions up to 250 mm in diameter, free of cracks, and second phase segregation.

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